Electron Spectroscopy Group at Physics Department, BNL

/ Peter Johnson, Alexei Fedorov, Tonica Valla/

Angle resolved photoemission:

- ✓ **High temperature superconductors /$Bi_2Sr_2CaCu_2O_8$, Sr_2RuO_4 /**
- ✓ **Low-dimensional conductors /CDW, non-Fermi liquid behavior/**
- ✓ **Two-dimensional conductors /surface states, 2H-$TaSe_2$ /**
- ✓ **Amorphous films /search for the Coulomb gap/**

Spin-polarized photoemission:

- ✓ **Micro-Mott detector connected to the Scienta analyzer /surface states in Gd(0001)/**

BROOKHAVEN NATIONAL LABORATORY

High resolution Angle Resolved Photoemission studies of Charge Density Wave materials: quasi-one-dimensional Blue Bronze $K_{0.3}MoO_3$ and two-dimensional dichalcogenide 2H-$TaSe_2$

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Why are we interested in high energy and momentum resolution? What are the goals?

A. Nesting properties of the Fermi surfaces

/Charge density waves/

B. Photoemission spectral functions A(k,ω)

/direct comparison with theoretical predictions/

Outline

Experimental details:

- ✓ Angle resolved photoemission
- ✓ Photoelectron spectrometer

$K_{0.3}MoO_3$:

- ✓ Crystal structure
- ✓ Electronic structure
- ✓ Structural studies /Charge Density Waves/
- ✓ Band structure of $K_{0.3}MoO_3$
- ✓ Fermi wave vectors versus temperature
- ✓ Incommensurate to commensurate CDW transition
- ✓ Signatures of non-Fermi liquid behavior

2H-$TaSe_2$:

- ✓ CDW, Nesting: “conventional” vs. “saddle point”
- ✓ Band mapping
- ✓ Coupling of quasiparticles to collective excitations
- ✓ Is 2H-$TaSe_2$ similar to the HTSC?

Angle Rsolved Photoemission

/band structure mapping/

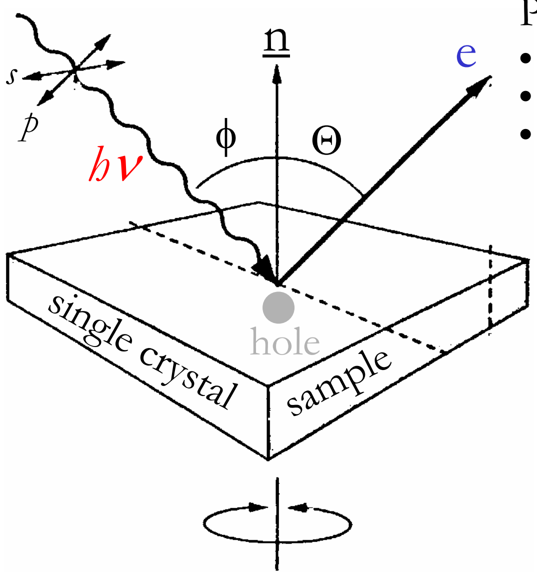
Experiment

$$k_{//} = \sin\Theta \times \sqrt{2 \times m_e \times \eta^2 \times (h\nu - \Phi - E_{binding})}$$

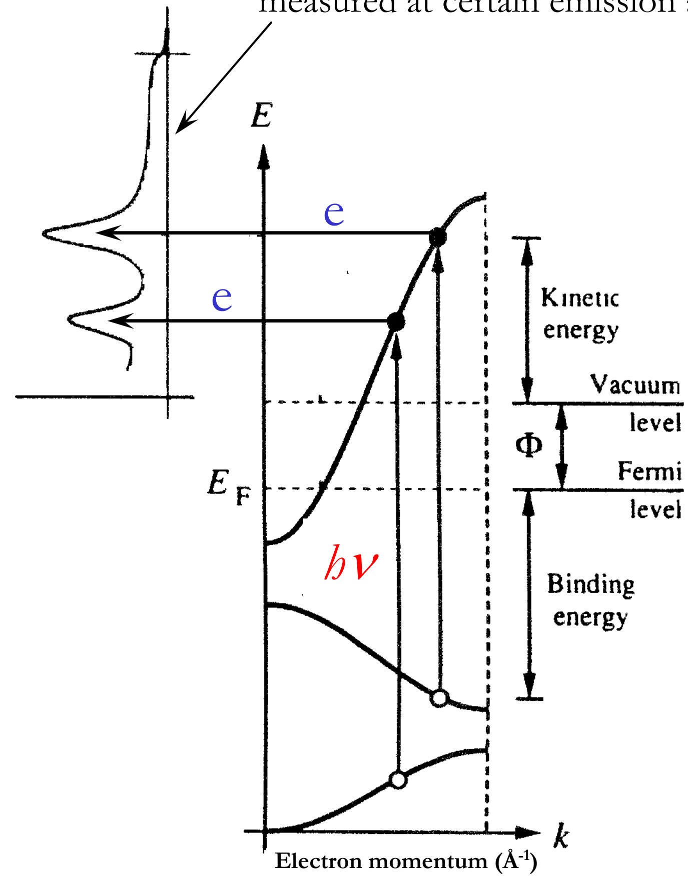
Data

Excitation Radiation
- photon energy
- polarization
- angle of incidence

Photoelectrons
- kinetic energy
- emission angle
- polarization



Energy Distribution Curves (photocurrent vs. kinetic energy) measured at certain emission angle

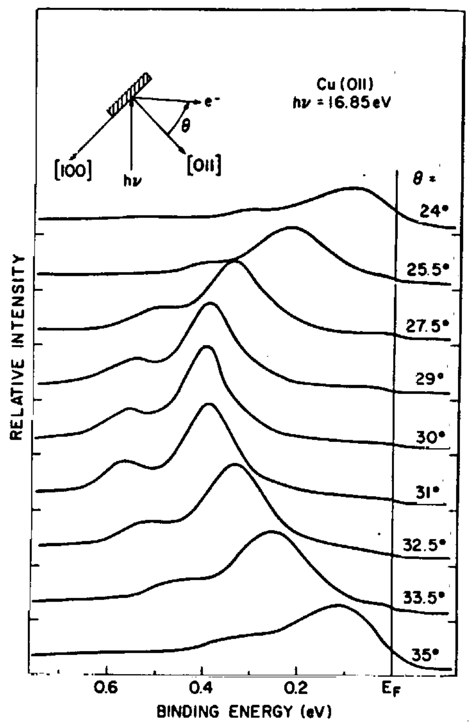


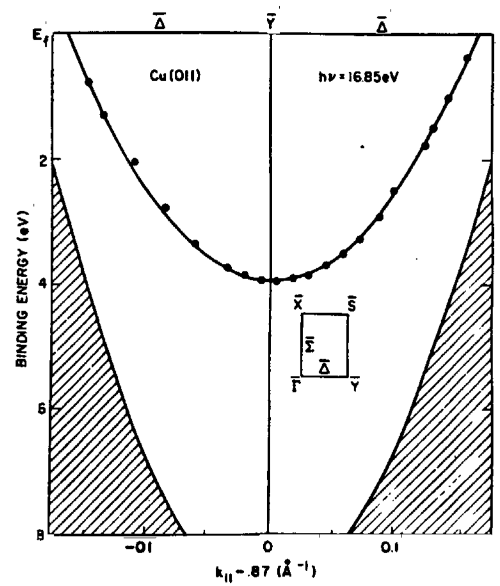
Important parameters:

Energy resolution (~20 meV)

Angular resolution (~2°)

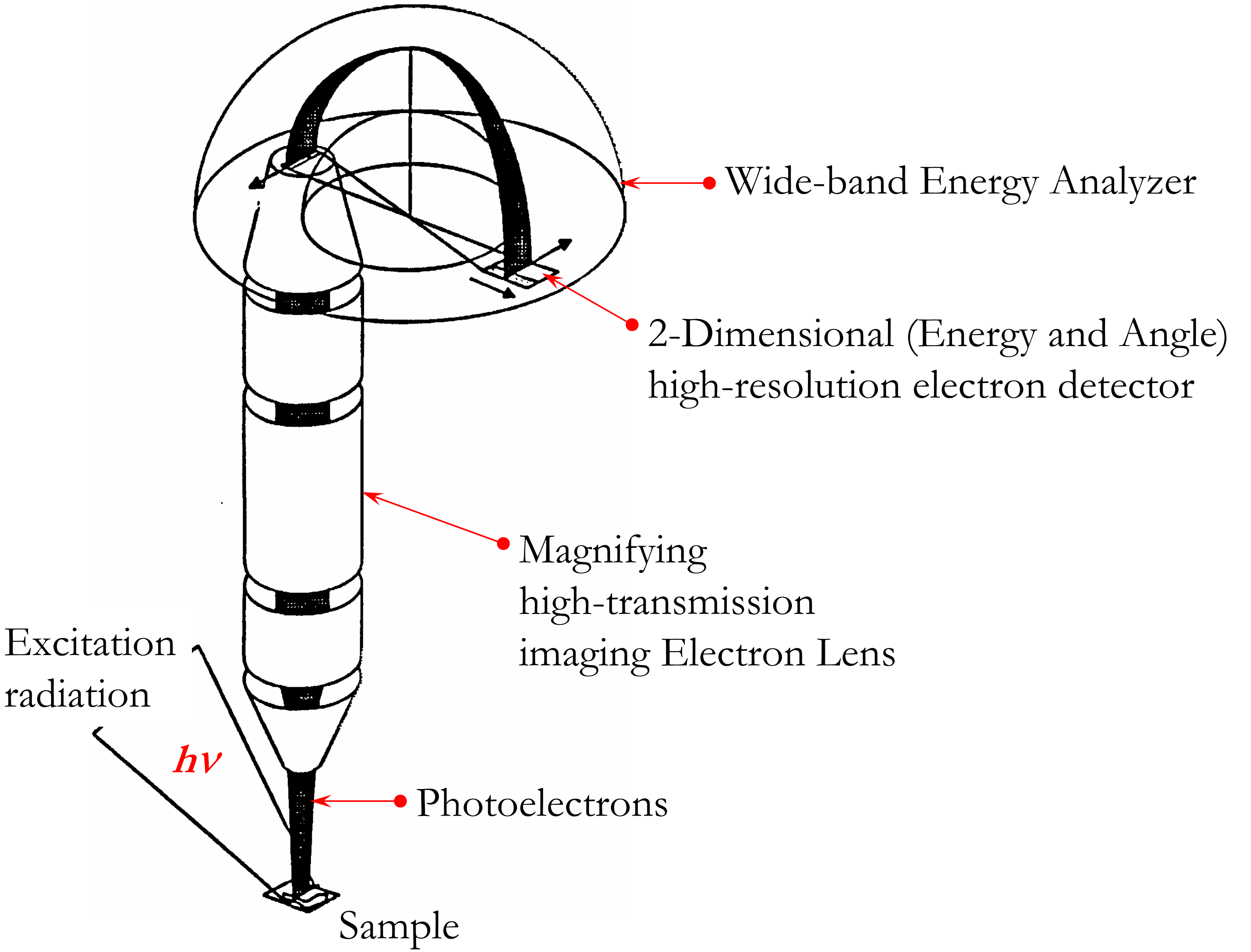
Surface State in Cu(011) mapped with ARPES
/S. Kevan, PRB **28**, 4822 (1983)/





New Instrumentation

/multi-channel detection in emission angle and kinetic energy/



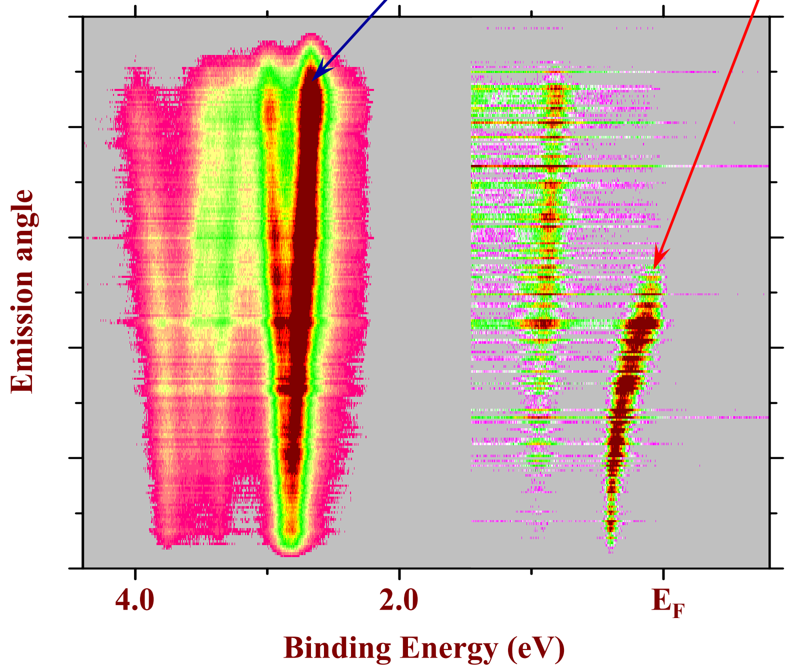
Photoelectron Spectrometer

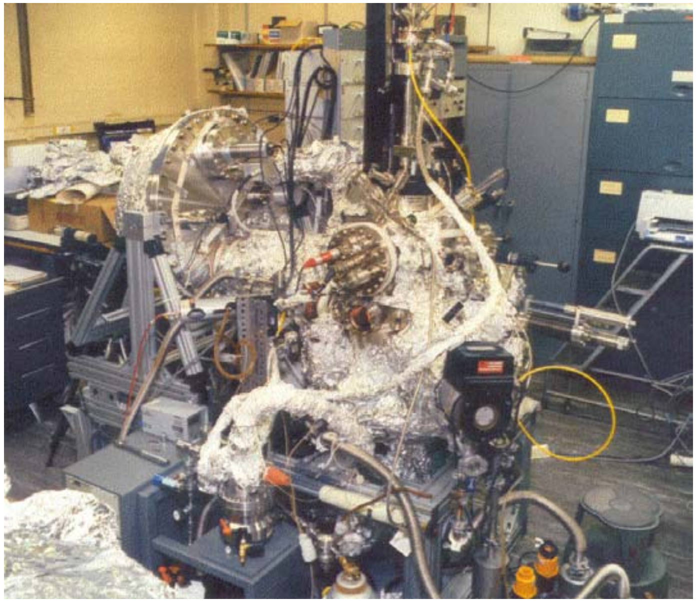
SES-200: 200 millimeters hemispherical deflector capable of multichannel detection in emission angle and kinetic energy

Example of angle resolved data:

hν = 21.22 eV /He I radiation/

Cu(111), bulk bands ● and sp-surface state ●



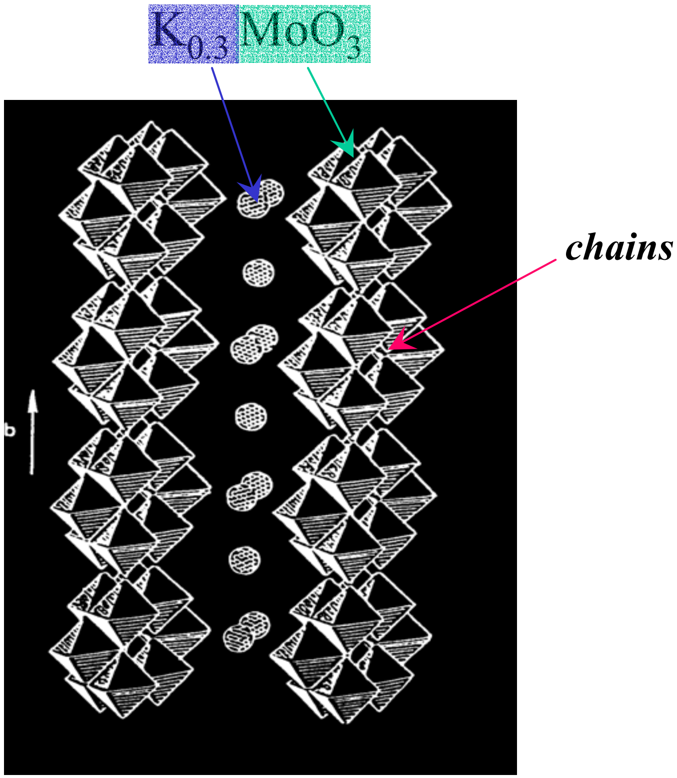


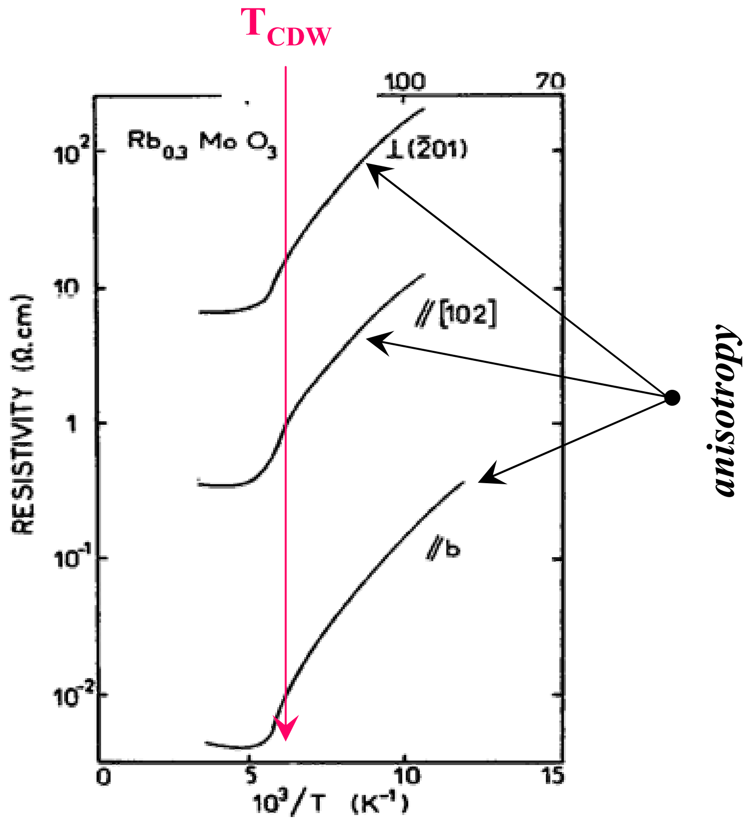
- ✓ **Energy resolution ~ 10 meV**
- ✓ **Angle resolution ~ 0.2 °**
- ✓ **Base pressure ~ 2× 10^{-11} Torr**

Presently located at the undulator beamline U13UB at the National Synchrotron Light Source

Why are we interested in Low-dimensional materials?

- Charge Density Waves (CDW) /Peierls transitions/
- Electron correlation effects /non-Fermi liquid behavior, spin-charge separation, HTSC/

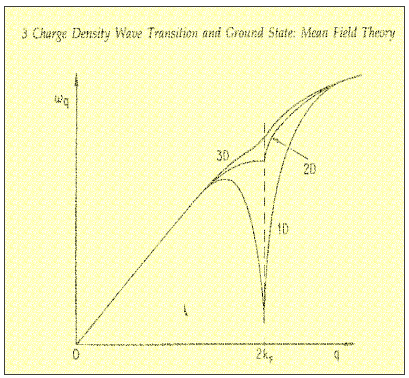




J.-P. Pouget et al., J. Physique Lett. 44, L113 (1973)

Charge Density Waves

/ Density Waves in Solids, G. Grüner, Addison Wesley, 1994 /



3.1 The Kohn Anomaly and the Peierls Transition: Mean Field Theory

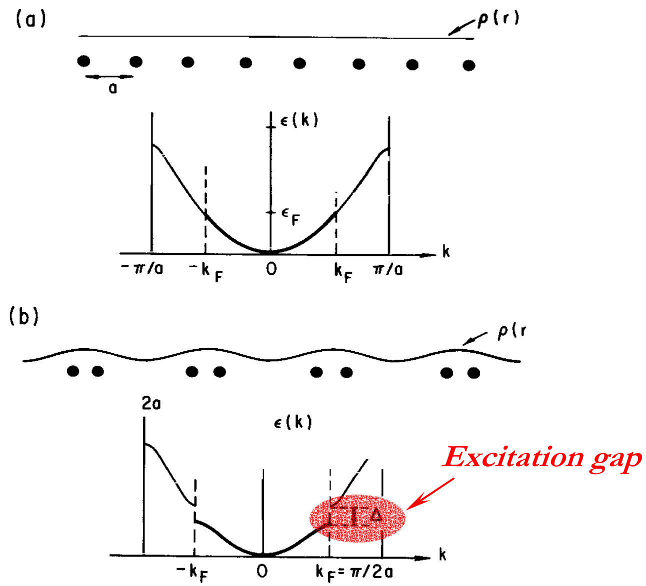
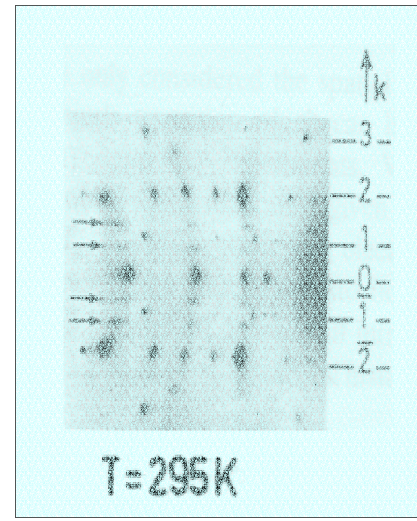


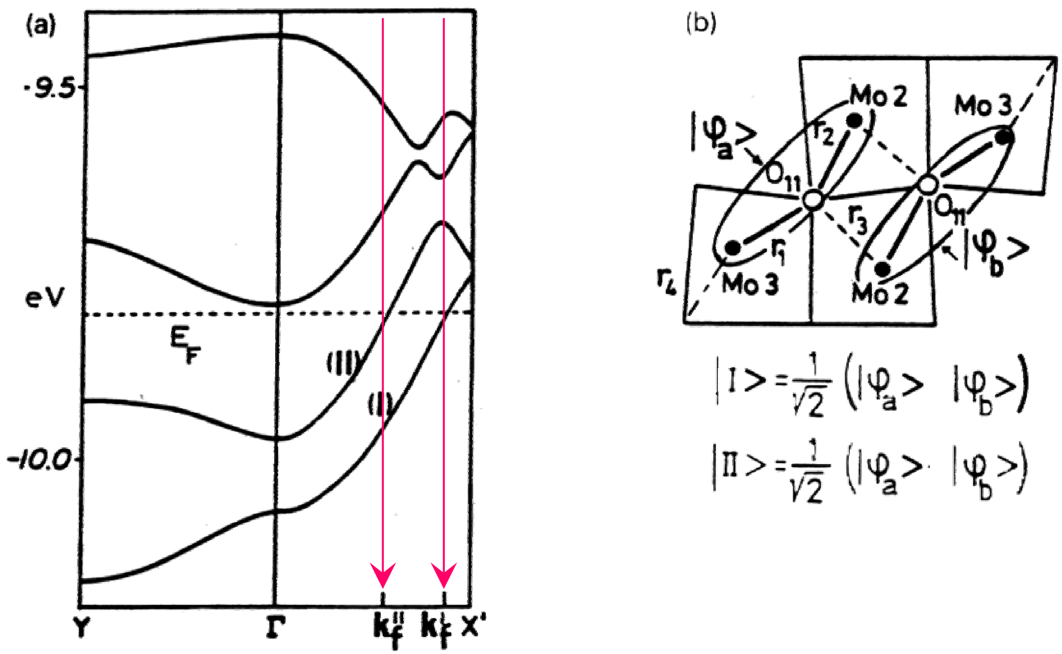
Figure 3.5. The single particle band, electron density, and lattice distortion in the metallic state above T_{CDW} and in the charge density wave state at $T = 0$. The figure is appropriate for a half-filled band.



Electronic structure of $K_{0.3}MoO_3$

/tight-binding calculations/

M.-H. Whangbo and L.F. Schneemeyer, Inor. Chem. 25,2424 (1986)



Two bands crossing the Fermi level

How many Charge Density Waves?

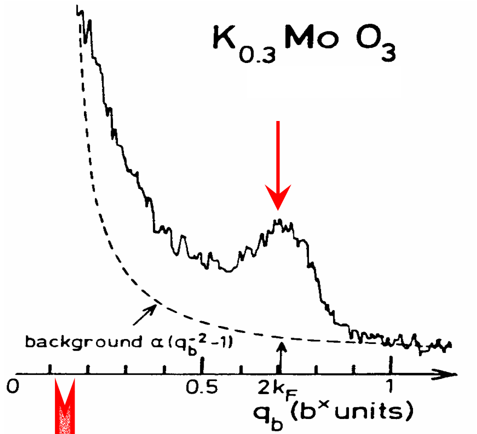
Structural studies of CDW in $K_{0.3}MoO_3$

/Single Charge Density Wave/

A. Diffuse X-ray scattering

$/ q_{CDW} = 2k_F b^{\times} /$

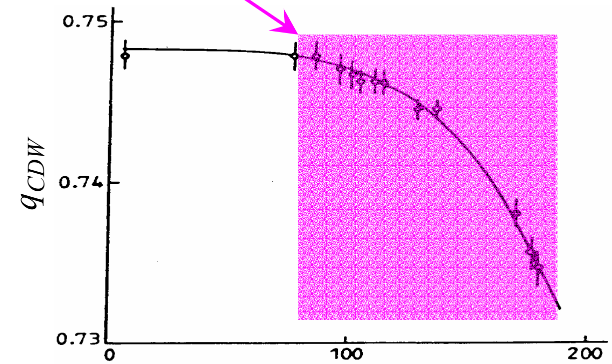
J.-P. Pougetet al.



B. Temperature dependent neutron scattering

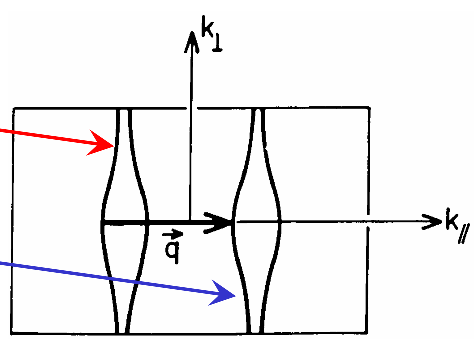
/incommensurate to commensurate transition/

M.Sato, H. Fujishita and S.Hoshito,
J. Phys. C: Solid State phys., 16, L877 (1983)



Nesting:

Fermi surface of the first band is nested to the Fermi surface of the second band



CDW wave vector
$q_{CDW} : k_{F1}+k_{F2}$

Temperature dependence of CDW wave vector:

◊ **Thermally activated charge transfer between bands crossing the Fermi level and third band above it** ***/Pouget et al./***

◊ **Shift of the chemical potential** ***/Pouget & Nougera, Artemenko et al./***

◊ **Hidden temperature dependence of the nesting vector** ***/Intention of the present study/***

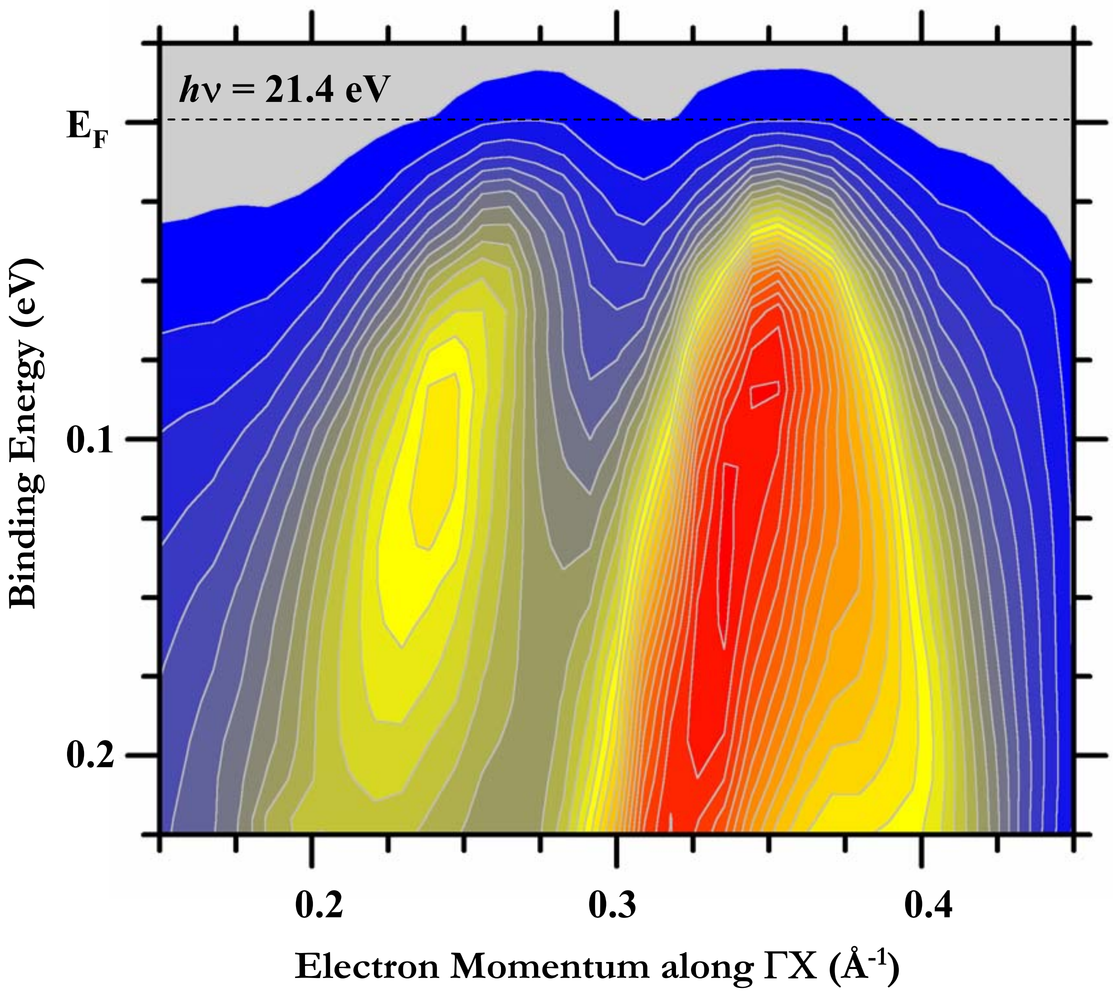
Goals of photoemission experiment:

◊ **Direct monitoring k_{F1} and k_{F2}**

◊ **Temperature dependence of $(k_{F1}+k_{F2})$**

Direct monitoring electron bands in $K_{0.3}MoO_3$

/3-D maps of photocurrent/



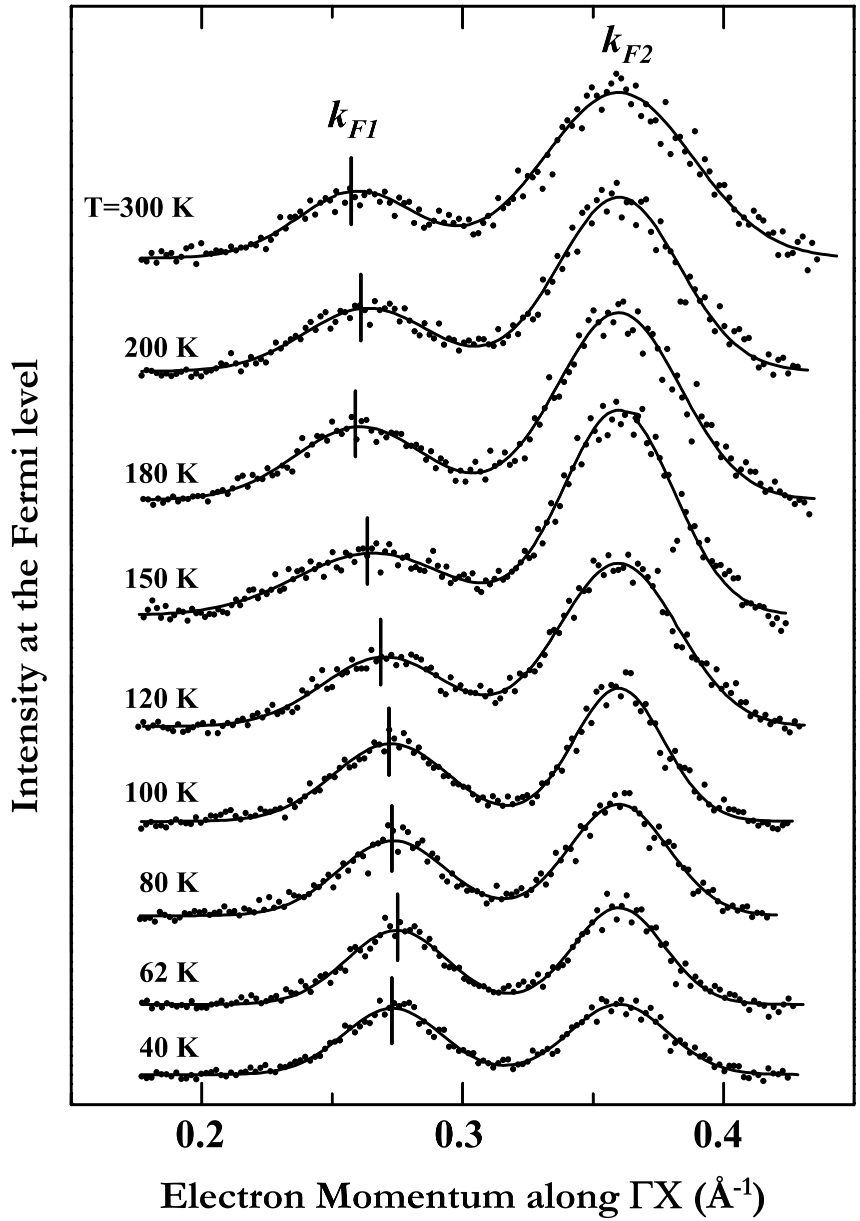
<u>Experimental details:</u>

Samples cleaved *in situ*

Liquid He cryostat provides temperatures from ~20 K to ~450 K

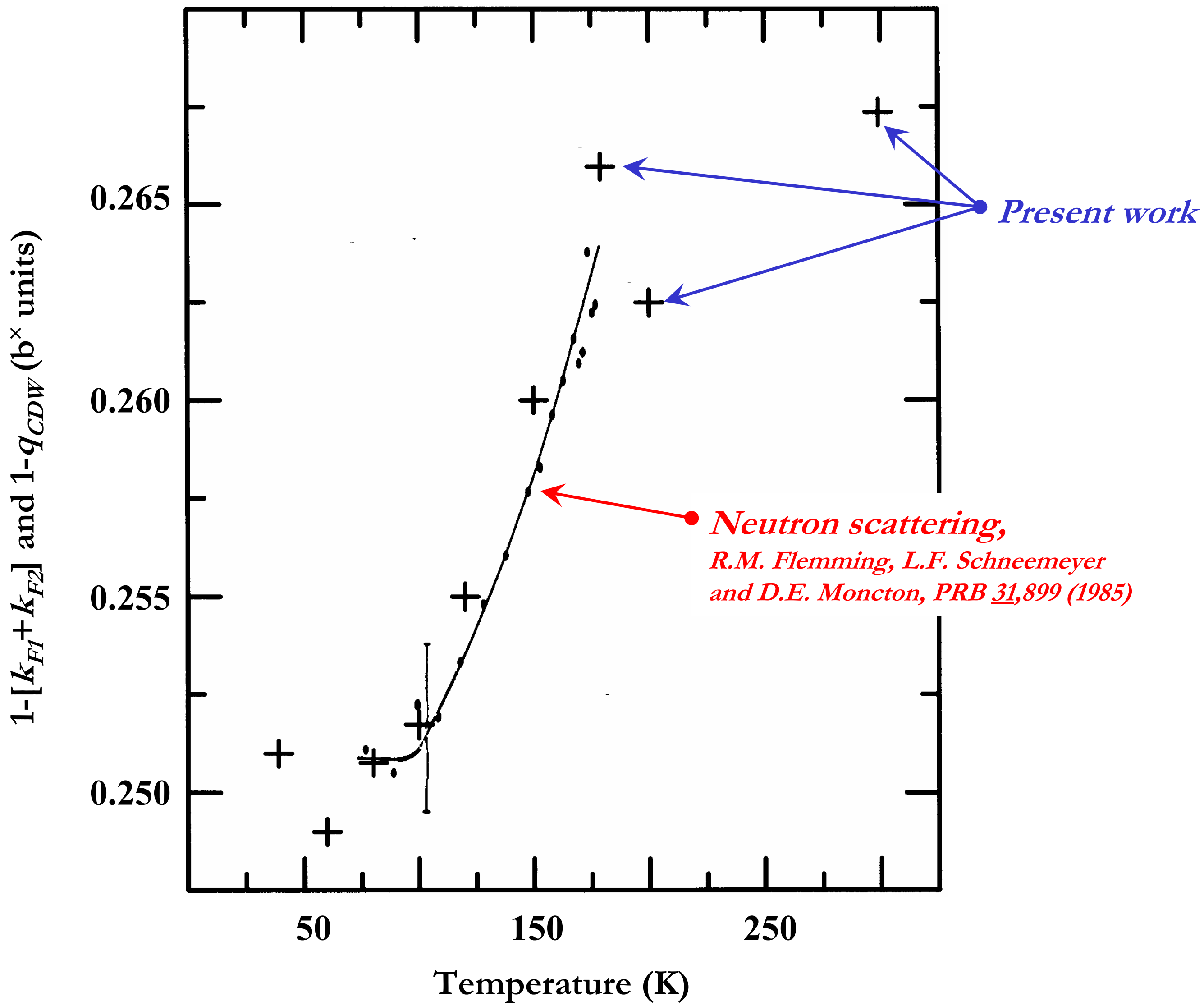
Temperature monitored with OMEGA CY7 sensor

Momentum Distribution Curves at E_F



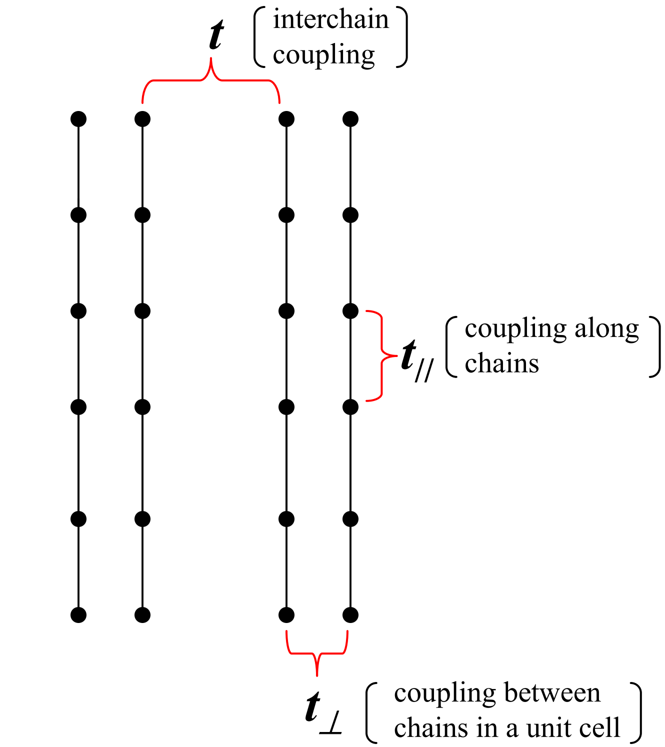
Incommensurate to commensurate CDW transition in $K_{0.3}MoO_3$

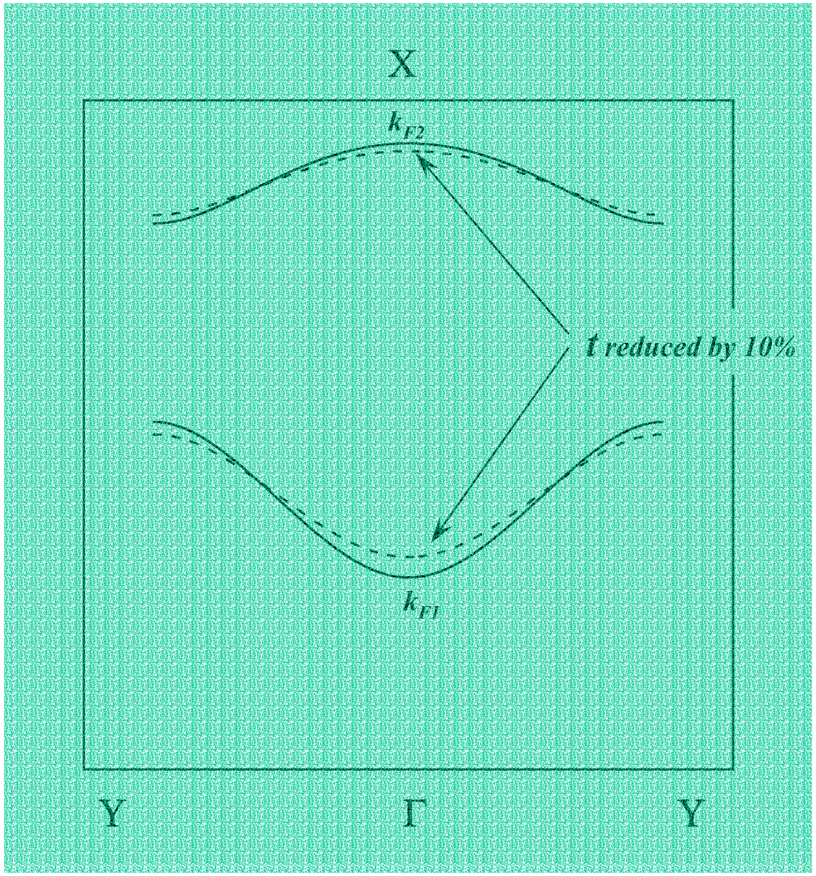
/comparing neutron scattering data with nesting vector measured in photoemission experiment/



Fermi surface of an array of coupled chains

/tight binding calculation/





<u>*Fermi surface is given by:*</u>

$$\mu = -2\cos(k_{//}) \pm (t_{\perp} + 2t_{\perp}\ t\cos(k_{\perp}) + t)$$

What are the signatures of non-Fermi liquid behavior in photoemission?

Spin-charge separation $\Rightarrow$ Observation of two dispersing features corresponding to the charge and spin degrees of freedom

Breakdown of the quasiparticle picture $\Rightarrow$ Suppression of the spectral weight at the Fermi energy

J. Phys.: Condens. Matter 5 (1993) 8305–8336. Printed in the UK

Charge–spin separation and the spectral properties of Luttinger liquids

Johannes Voit
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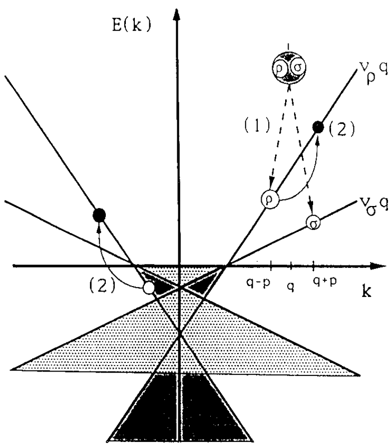
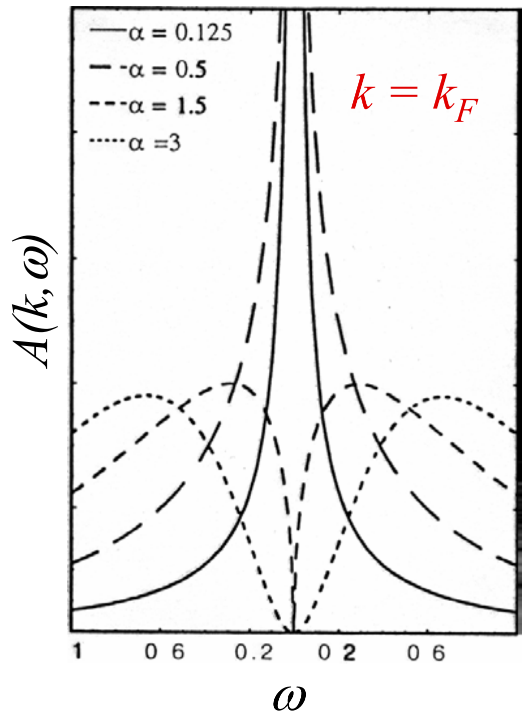
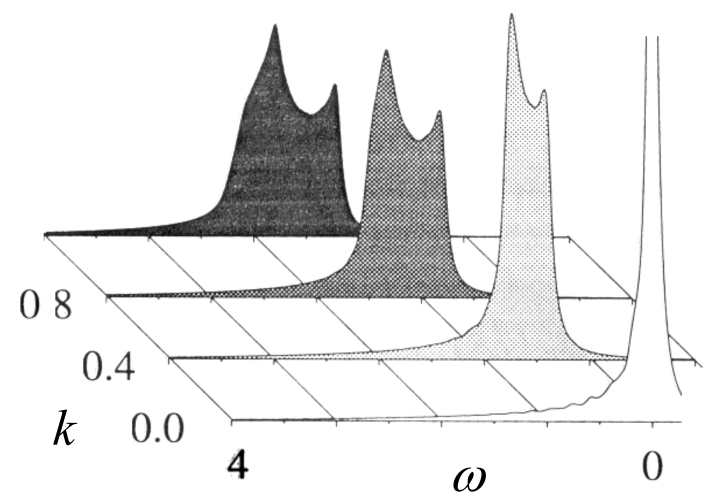




VOLUME 74, NUMBER 15 PHYSICAL REVIEW LETTERS 10 APRIL 1995

Anomalous Scaling and Spin-Charge Separation in Coupled Chains

Peter Kopietz, Volker Meden, and Kurt Schönhammer
Institut für Theoretische Physik der Universität Göttingen, Bunsenstrasse 9, D-37073 Göttingen, Germany
(Received 19 August 1994)

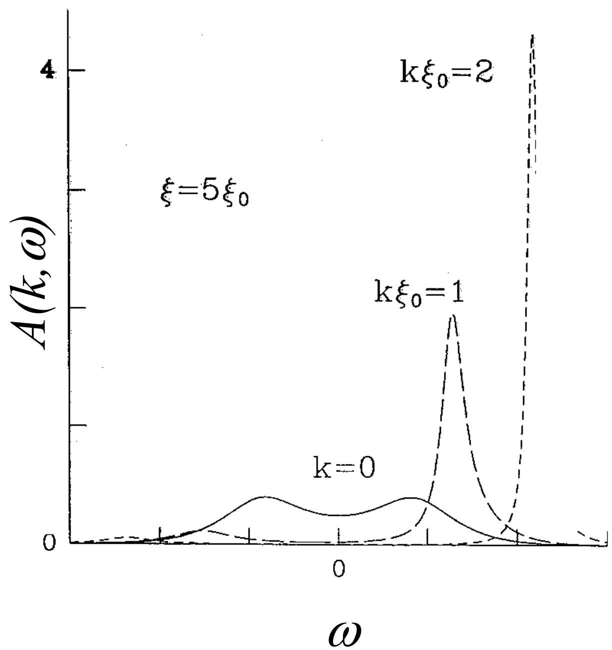


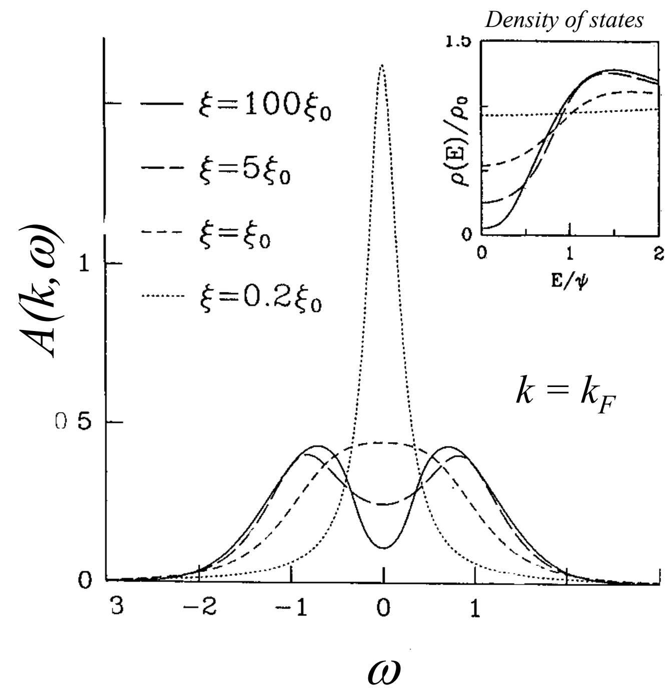
PHYSICAL REVIEW B VOLUME 54, NUMBER 18 1 NOVEMBER 1996-II
RAPID COMMUNICATIO

Non-Fermi-liquid behavior due to short-range order

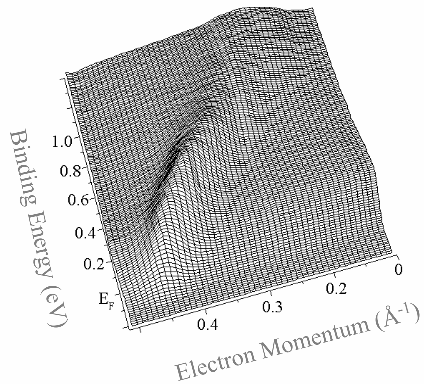
Ross H. McKenzie* and David Scarratt
School of Physics, University of New South Wales, Sydney 2052, Australia

(Received 23 May 1996)

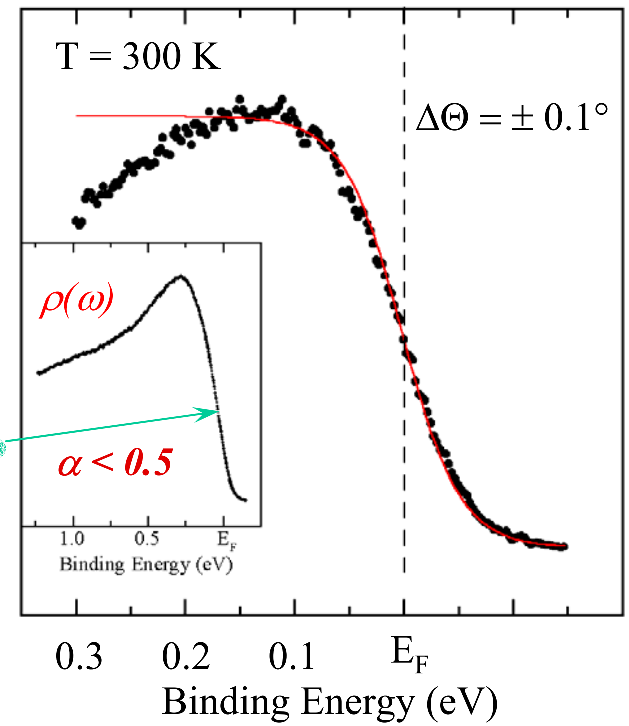




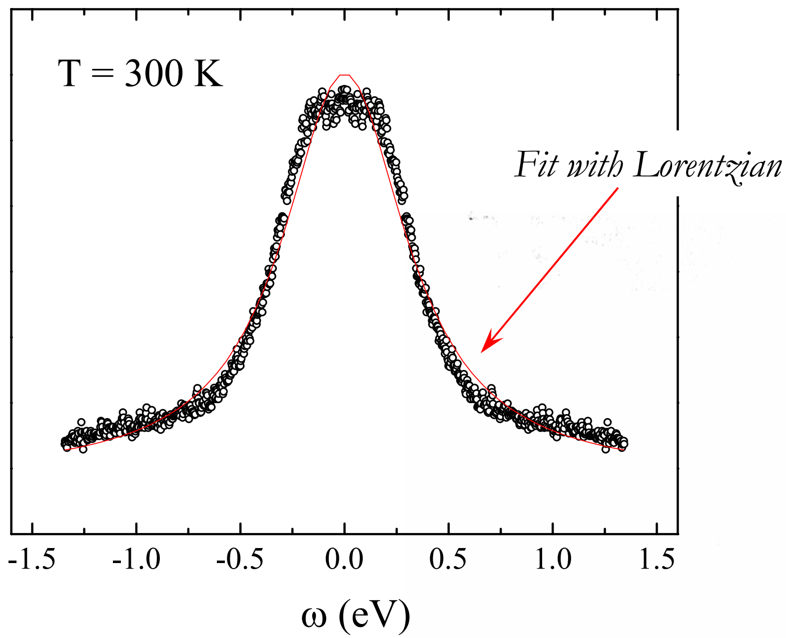
Spectral function in $K_{0.3}MoO_3$ at the Fermi wave vector



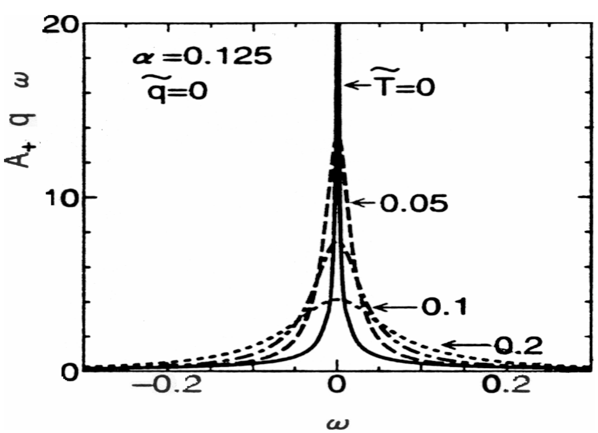
$\rho(\omega) \sim (\omega - E_F)^{\alpha}$



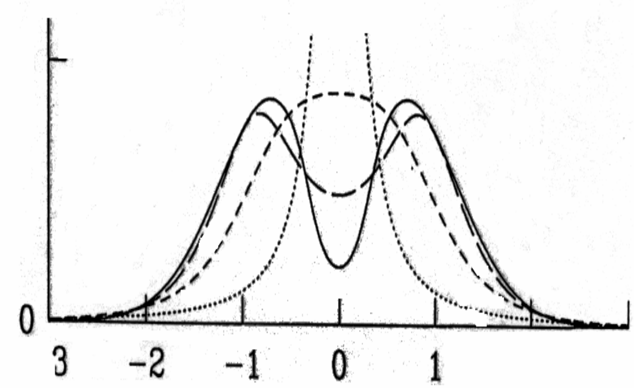
Spectral function symmetrized at k_F



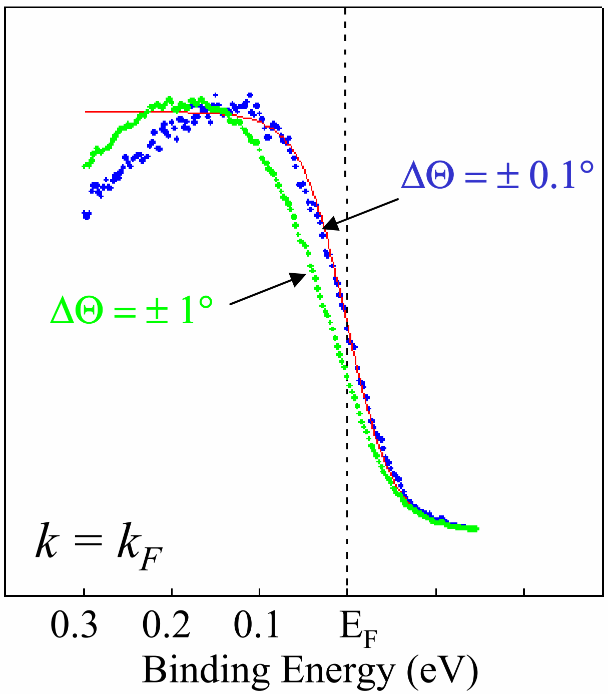
(a) Thermal fluctuations in Luttinger liquid



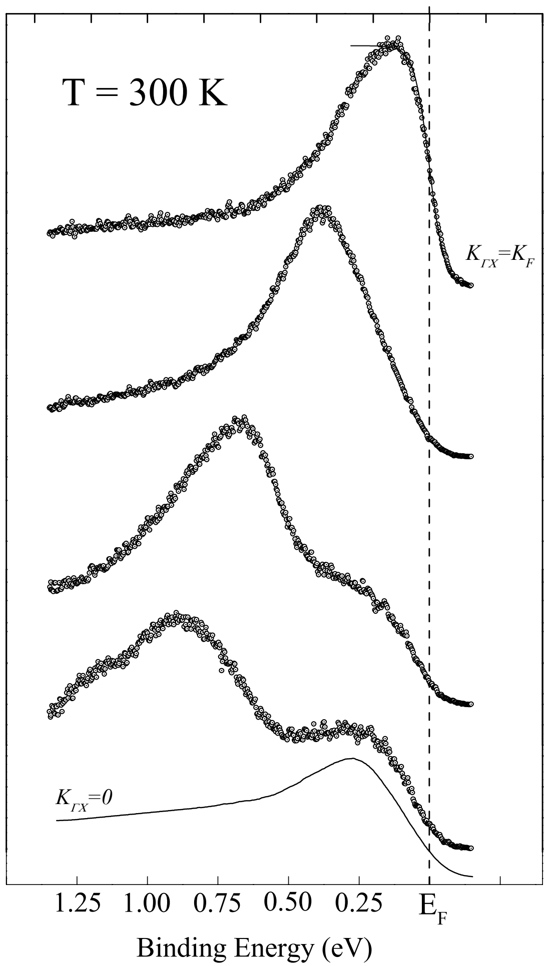
(b) CDW fluctuations

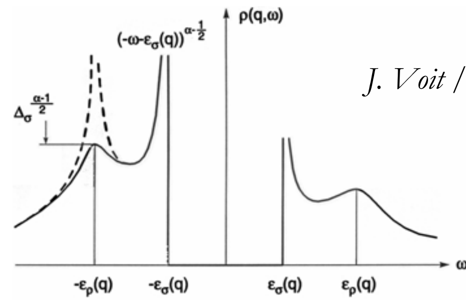


Suppression of spectral weight in photoemission from low-dimensional conductors: influence of momentum resolution

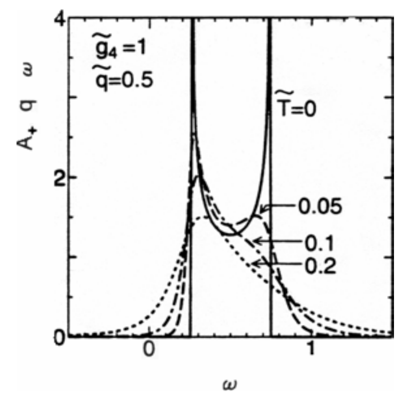


Spectral line-shapes versus electron momentum



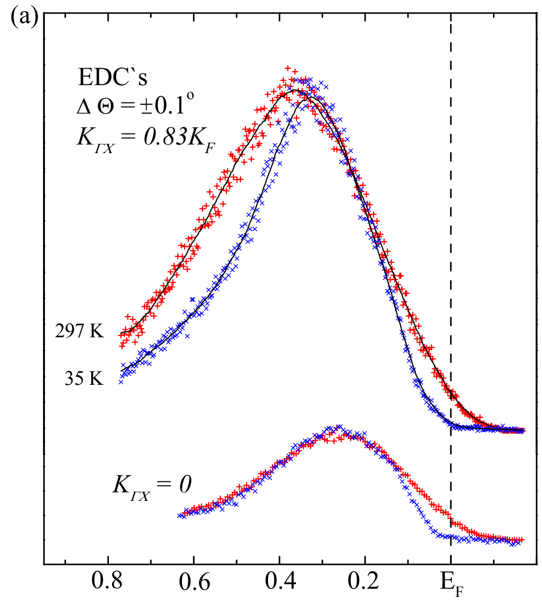


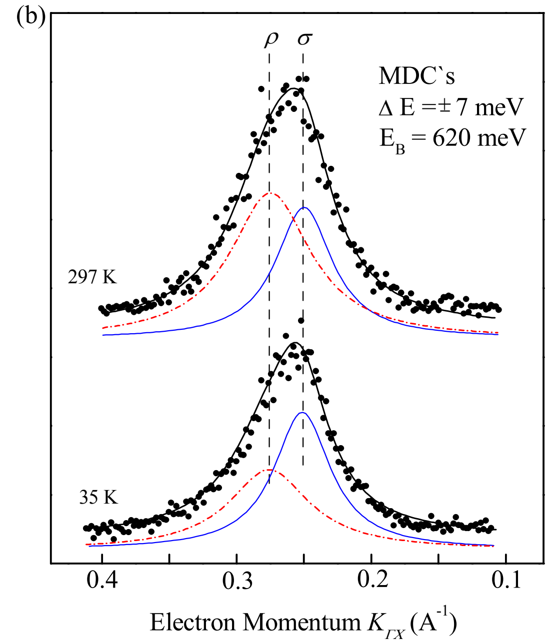
J. Voit /spin gap/

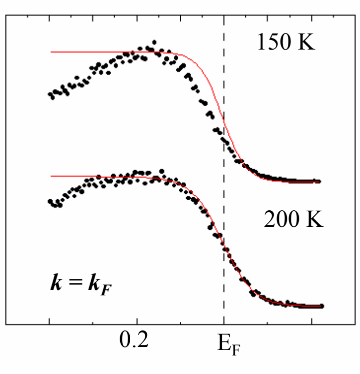


N. Nakamura & Y. Suzumura
/temperature effect/

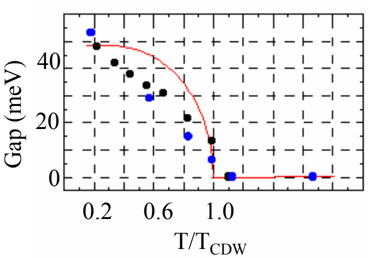
Spectral line-shapes versus temperature







Gap opening at the Fermi wave vector



$K_{0.3}MoO_3$, Summary:

Using ARP with high momentum reslution we have directly measured two Fermi wave vectors in quasi one-dimensional $K_{0.3}MoO_3$. By monitoring the temperature dependence of the Fermi wave vectors from 300 to 40 K, it was possible for the first time to compare the temperature dependence of the Fermi wave vectors and the CDW nesting vector. Our results show unambiguously that the temperature dependence of the CDW nesting vector observed in X-ray and neutron scattering experiments is primarily due to a change in the electronic structure.

We have demonstrated that the suppression of spectral weight at E_F seen in many one dimensional conductors could be an artifact of poor momentum resolution. The latter mimics effects arising from strong electron correlation. Hence, good momentum resolution is a *sine qua non* for studies of non Fermi liquid behavior. In the specific instance of $K_{0.3}MoO_3$, we did not find any suppression of intensity in our measurements of the spectral function. To our knowledge, this is the first time a Fermi edge has been observed in a quasi 1-D CDW material. Our results show that one cannot conclude about the presence of a pseudogap in this material, solely on the basis of the observed spectral intensity near E_F. This issue can only be resolved by a detailed study of the photoemission lineshapes above and below the Peierls transition.

$2H\text{-}TaSe_2$: Motivations and Questions

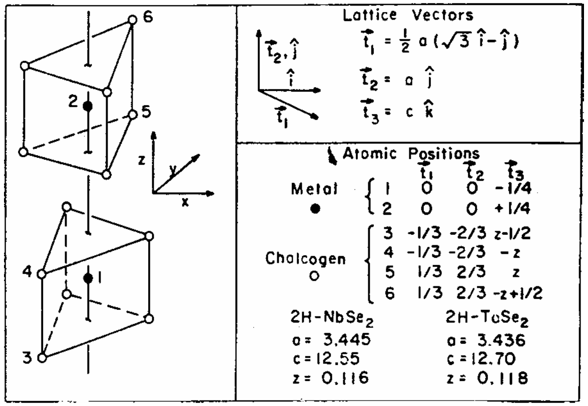
✓ **CDW coexists with superconductivity:**
$T_{CDW} \sim 122$ K ; $T_{SC} \sim 0.15$ K

✓ **What drives the CDW transition:**
"Conventional" Fermi surface nesting or "saddle point" nesting?

✓ **CDW does not remove the entire Fermi surface:** What happens to the excitations at the Fermi energy in a presence of the CDW gap?

2H Crystal structure

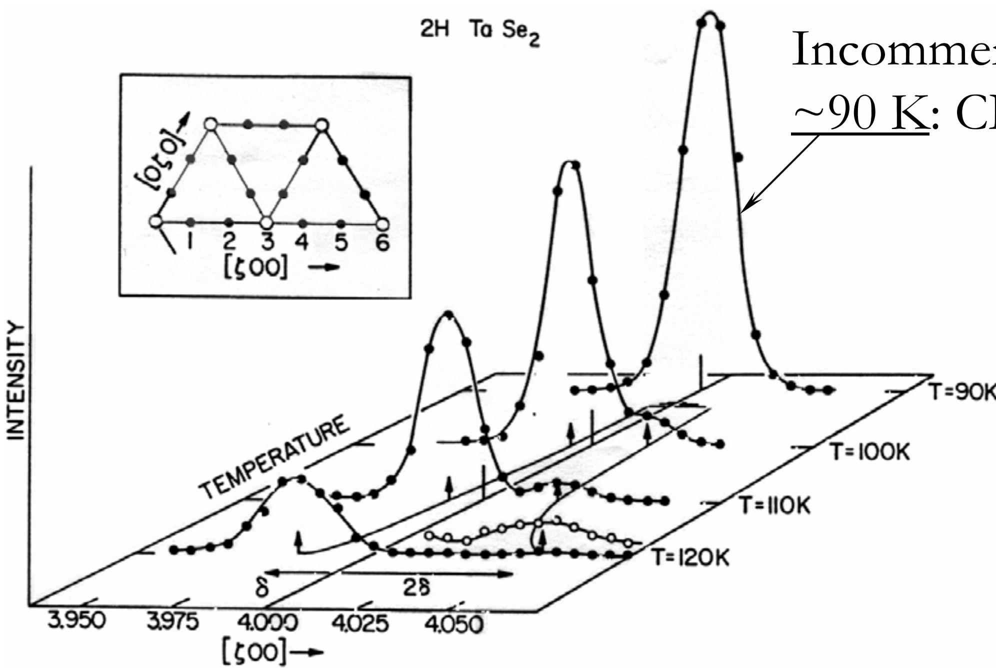
/D.E. Moncton, J.D. Axe, and F.J. DiSalvo, PRB **16**, 801(1977)/



Neutron scattering experiment

/superlattice due to the Charge Density Wave/

D.E. Moncton, J.D. Axe, and F.J. DiSalvo, PRL 34, 734 (1975)



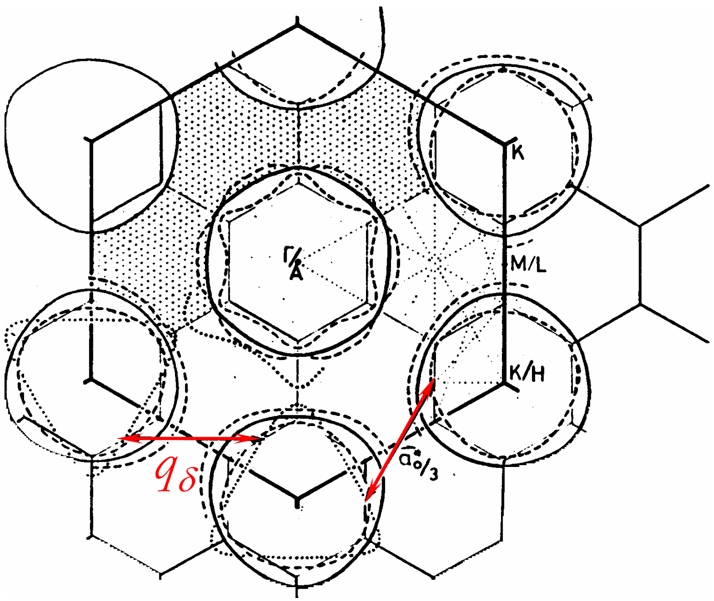
Incommensurate CDW: 122.3 K

~90 K: CDW locks to 3a superlattice

CDW wave vector: $q_\delta = 4\pi\,\{1\text{-}\delta(T)\}/a\sqrt{3}$

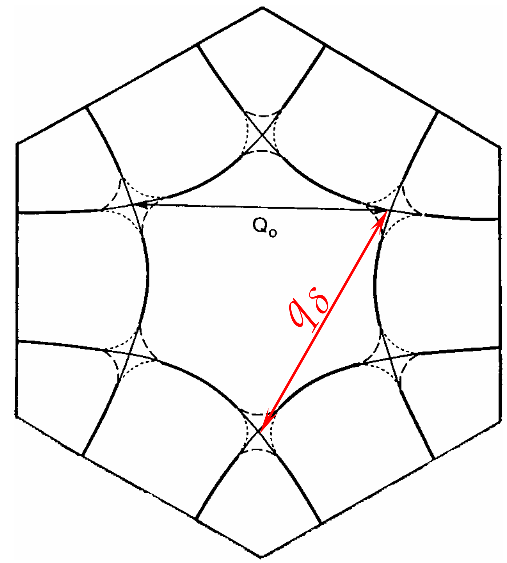
Nesting

A. Fermi surface nesting



J.A. Wilson, PRB 15, 5748 (1977)
G. Wexler and A.M. Wooley, J. Phys. C 9, 1185 (1976)
L.F. Mattheiss, PRB 8, 3719 (1973)

B. "Saddle point" nesting

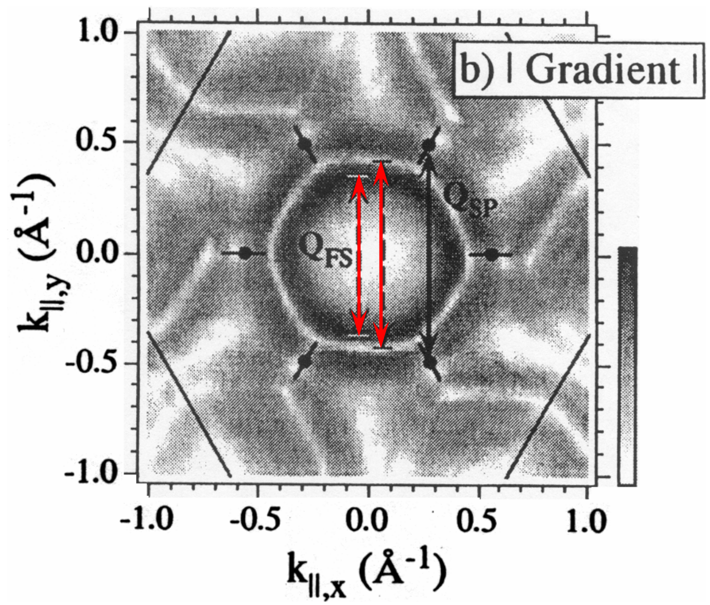


T.M. Rice and G.K. Scott, PRL 35, 120 (1975)

What is known?

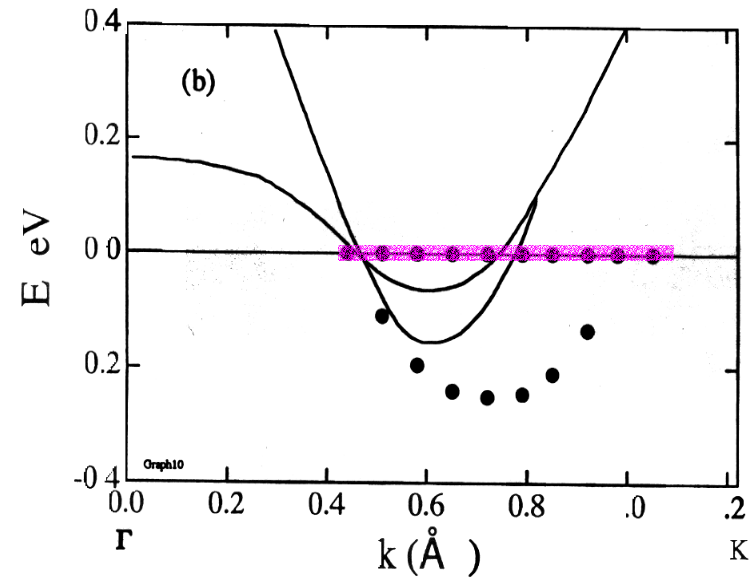
/ARPES studies/

A. "Regular" nesting



Th. Straub et al., PRL 82, 4504 (1999)

B. Saddle band ⇒ Rice-Scott model

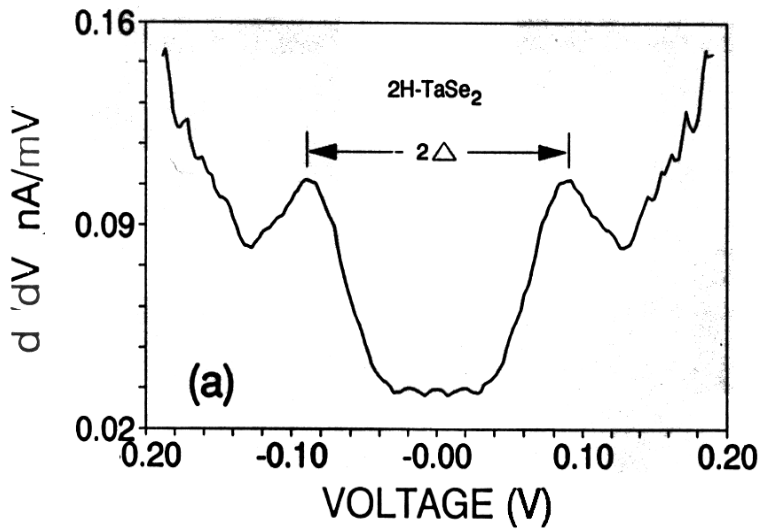


Rong Liu et al., PRL 80, 5762 (1998)

0.69 Å^{-1} < q_δ < 0.87 Å^{-1} ⇐ **Problems** ⇒ Saddle band, not a point

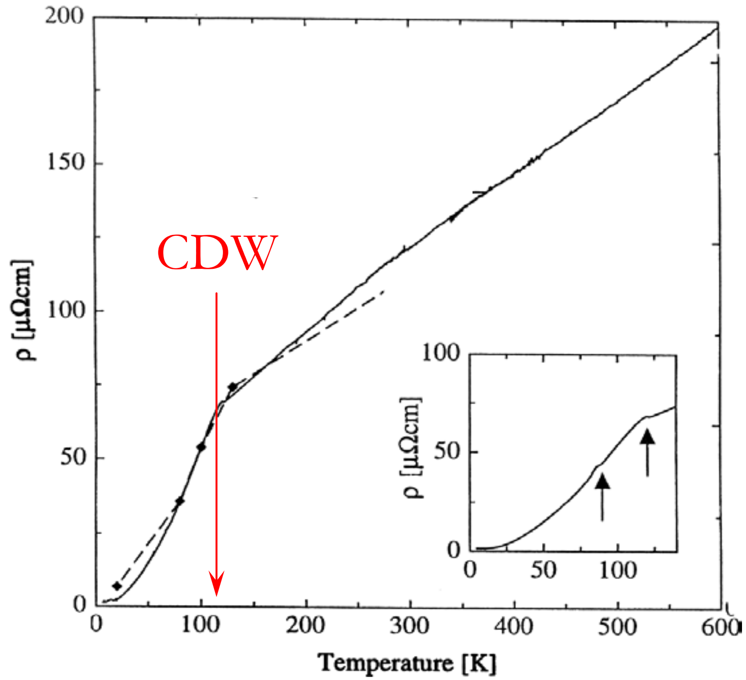
What does CDW do?

Opens up a gap, $2\Delta \sim 150$ meV
/STM data/



Z. Dai et al., PRB 48, 14543 (1993)

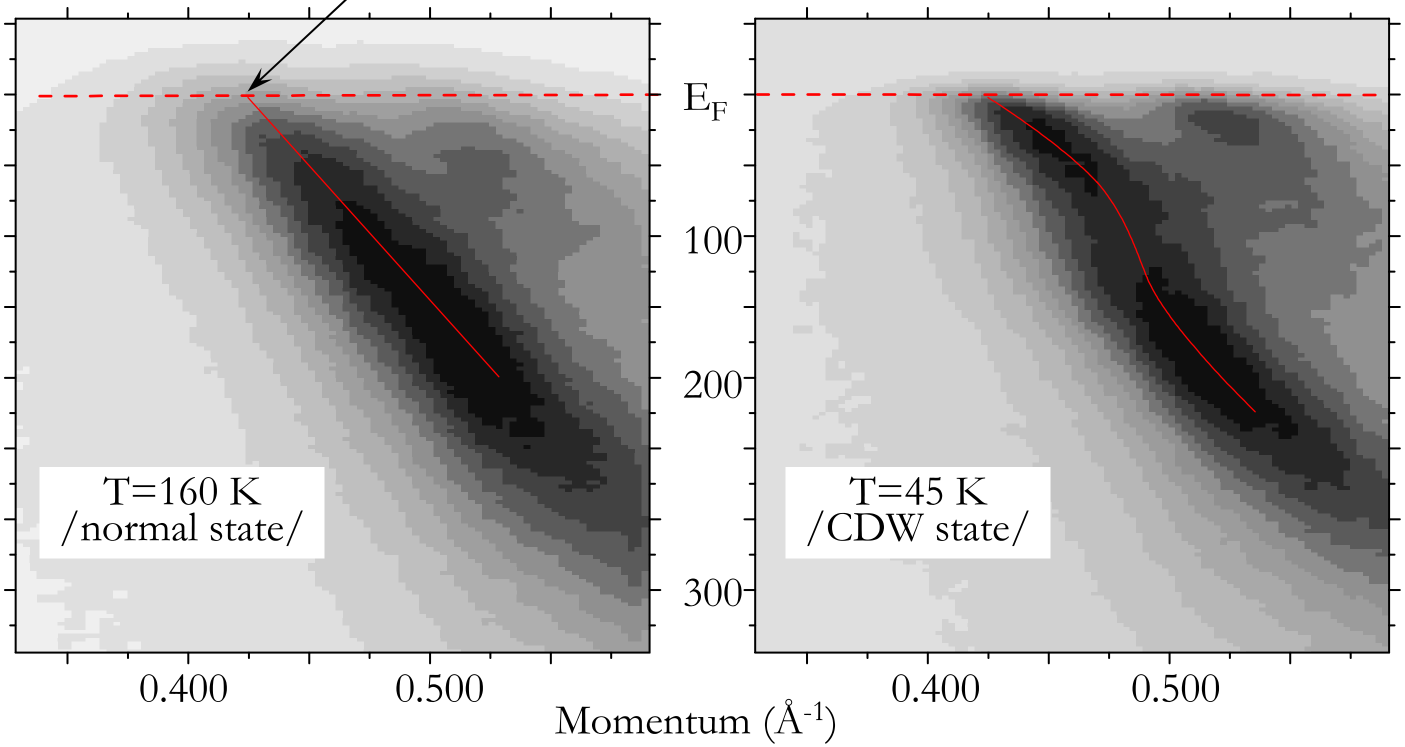
Freezes out scattering channels
/transport measurements/

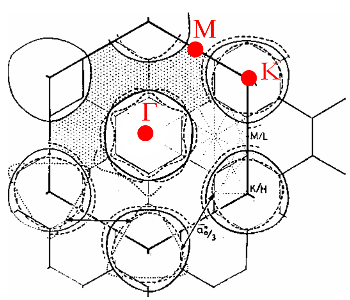


V. Vescoli et al., PRL 81, 453 (1998)

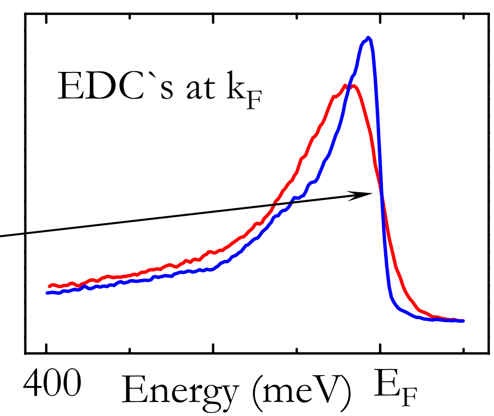
Band mapping along ΓM

Fermi level crossing: $k_F = 0.425$ Å^{-1}

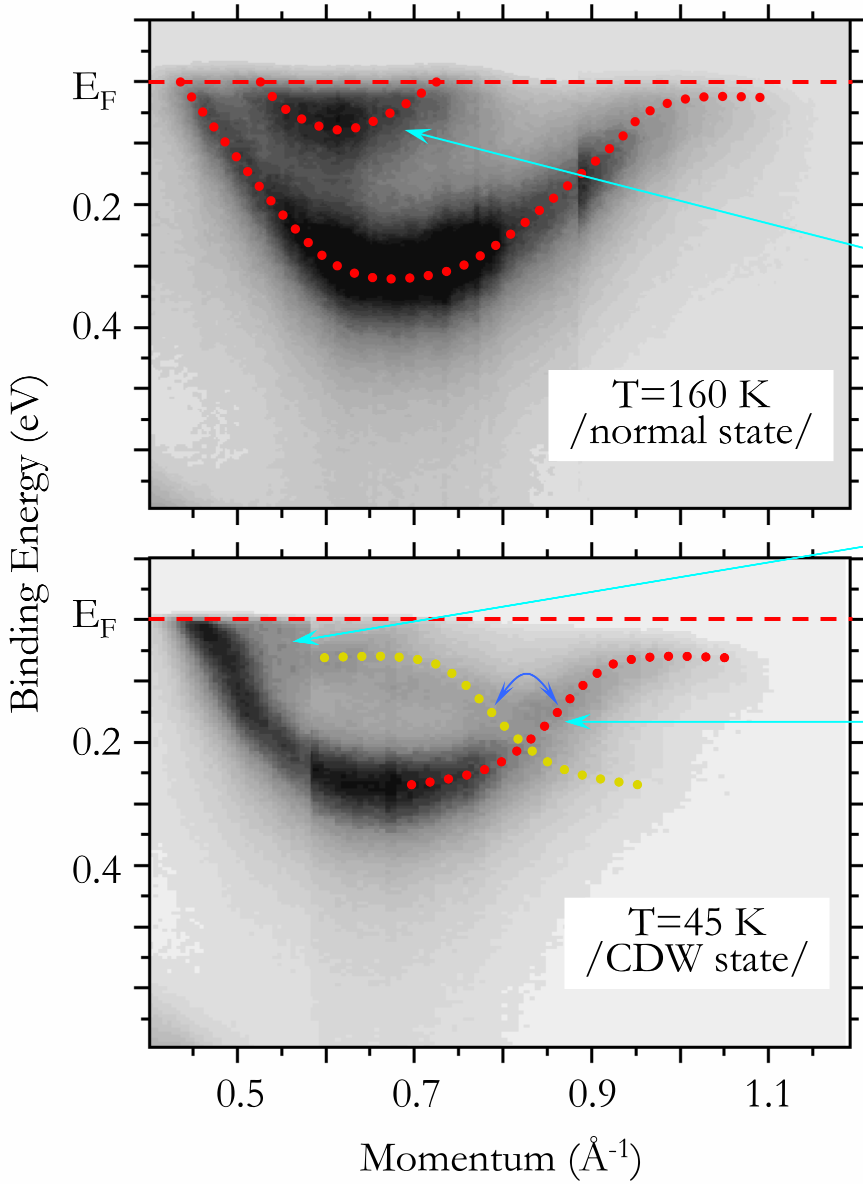




Nesting along ΓM
is not very good
and there is no gap
at the Fermi level...



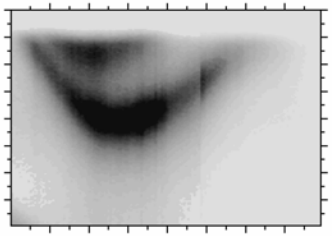
Band mapping along ΓK



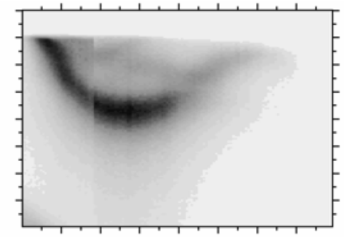
<u>New results:</u>

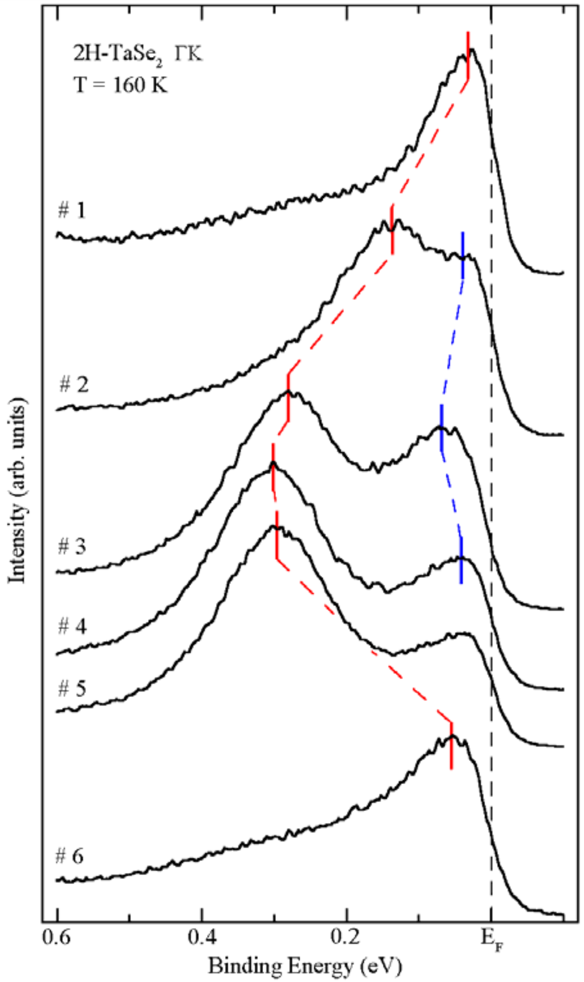
- Saddle point has a bandwidth of just ~50 meV and extends for only 0.2 Å^{-1}
- It is no longer there in the CDW-state
- Band "folds back" at ~0.825 Å^{-1}; This projects into ~2/3 of ΓM

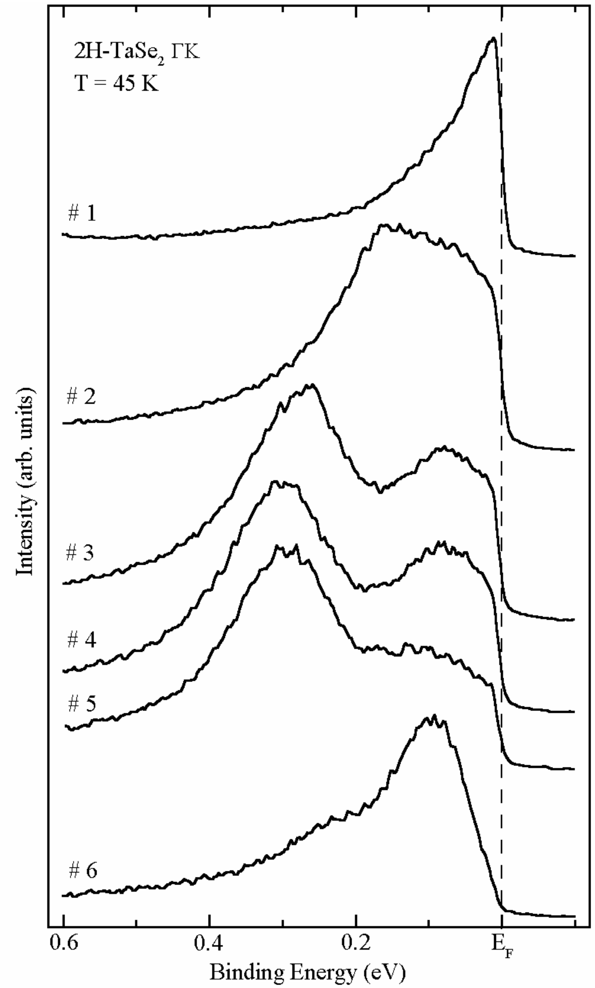
These observations point towards the Rice-Scott model



Energy distribution curves at few interesting points along ΓK

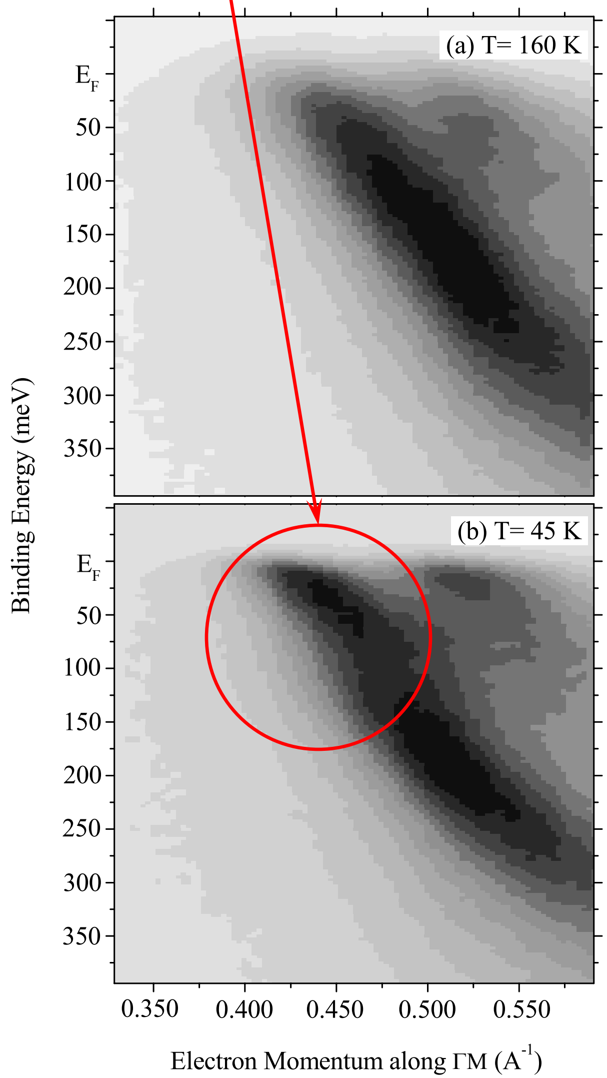


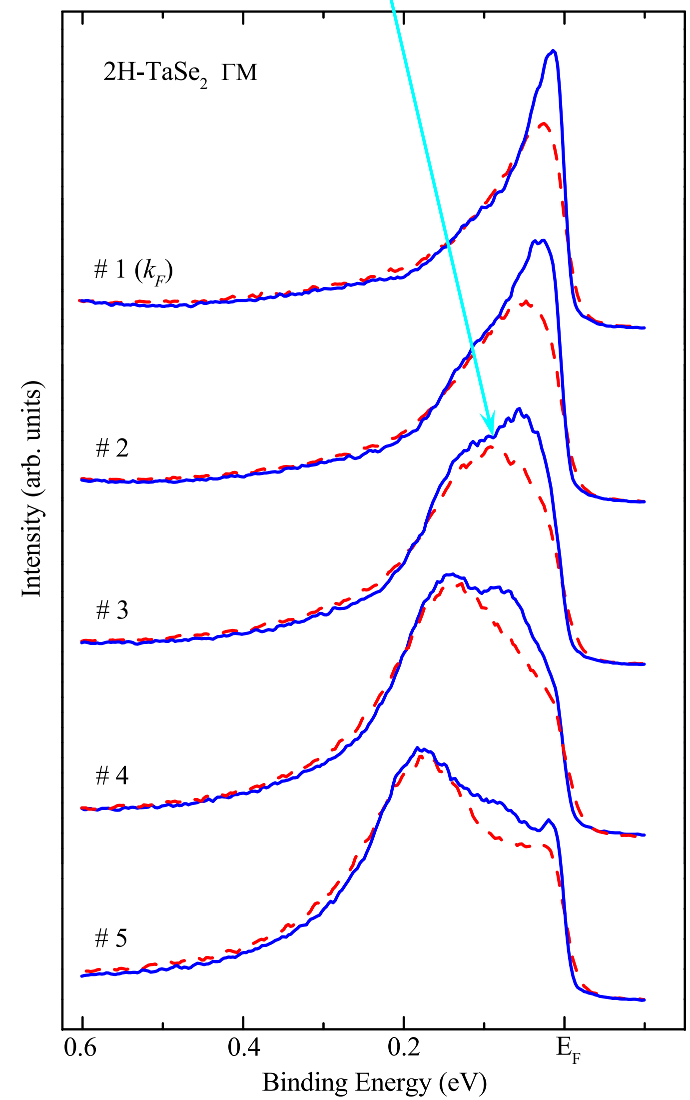




How does CDW affect low-energy excitations?

At **45 K** coupling of quasiparticles to the collective mode of some sort manifests itself via changes of both, ARPES **line-shapes** and **dispersion relations**

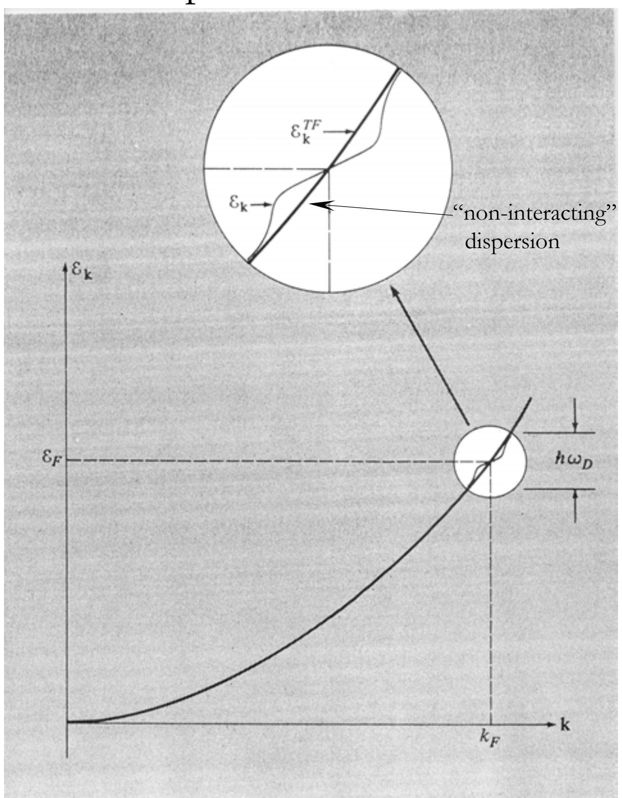




Electron-phonon coupling

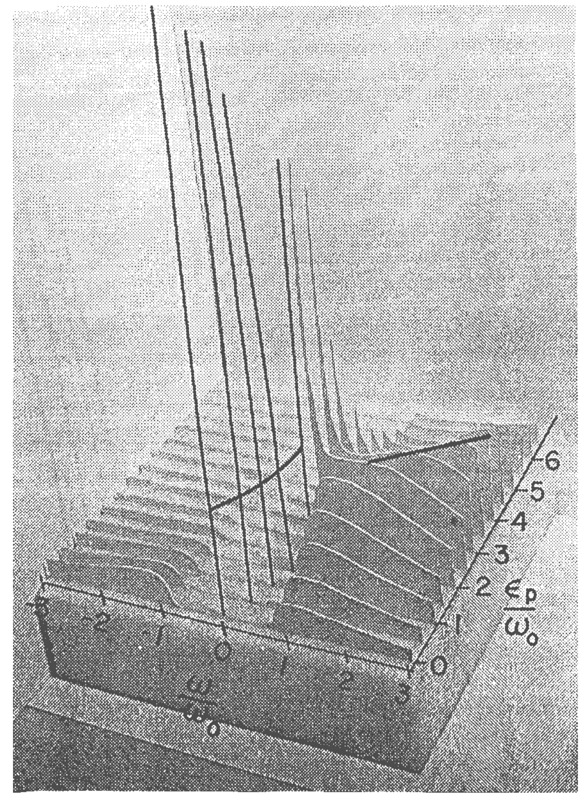
$$\textit{Spectral function: } A(k,\omega) \sim \frac{|\operatorname{Im}\Sigma(k,\omega)|}{[h\omega - e_k - \operatorname{Re}\Sigma(k,\omega)]^2 + \operatorname{Im}\Sigma(k,\omega)^2}$$

Dispersion relations



Solid State Physics
Neil W. Ashcroft
N. David Mermin

Spectral functions



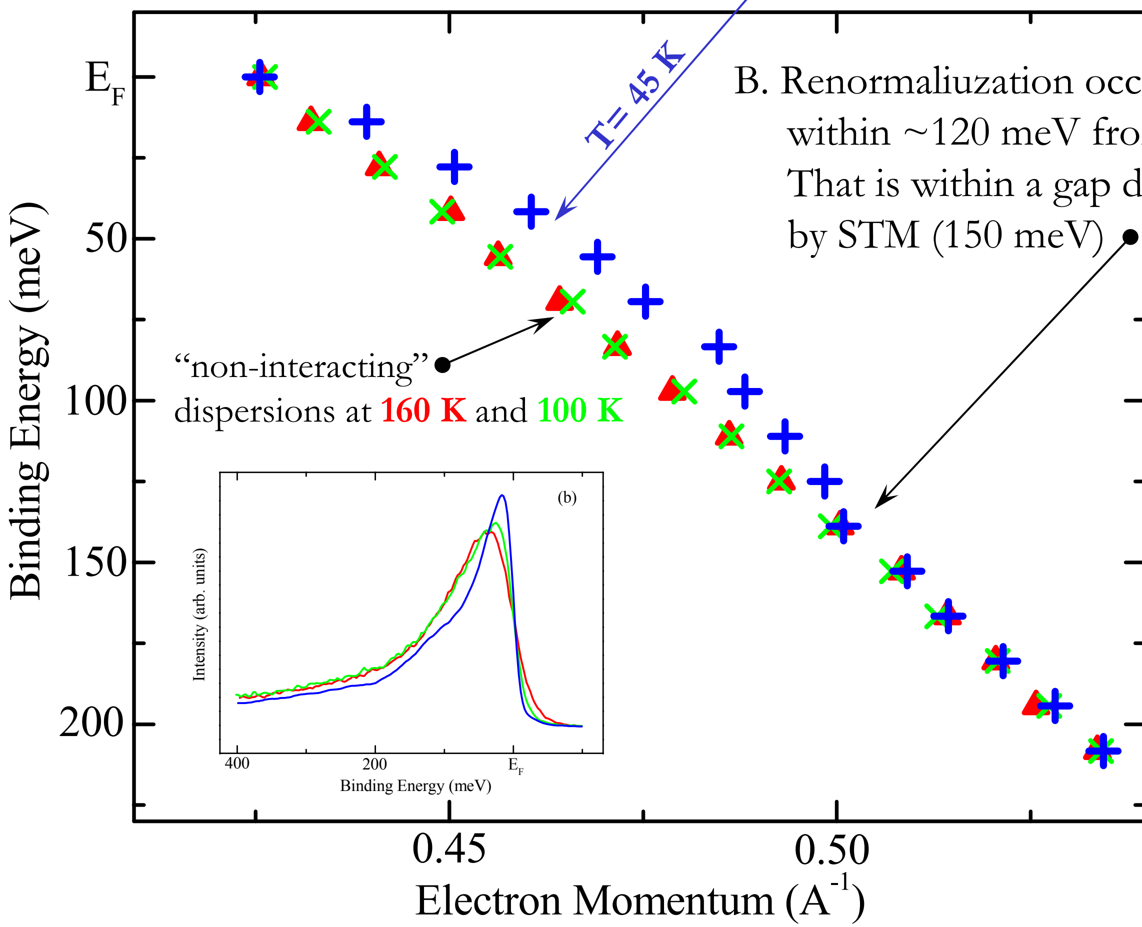
Douglas J. Scalapino
in Superconductivity,
R.D. Parks, editor

What is this collective mode?

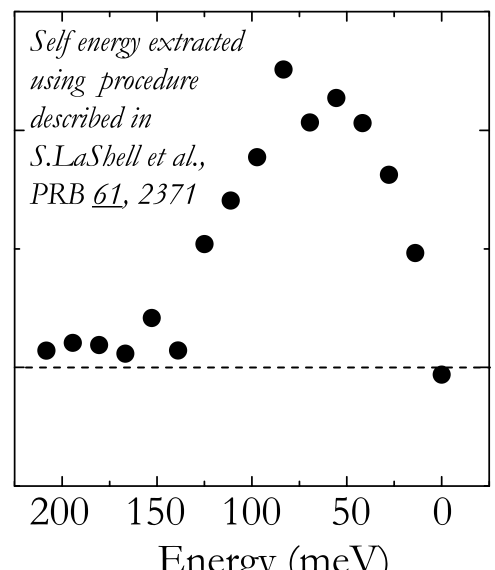
/a few clues from dispersion relations/

A. When CDW is commensurate with the lattice
"Renormalization" of dispersion becomes obvious

B. Renormaliuzation occurs within ~120 meV from E_F
That is within a gap detected by STM (150 meV)



C. Real part of the self energy peaks at ~80 meV, again within a CDW gap



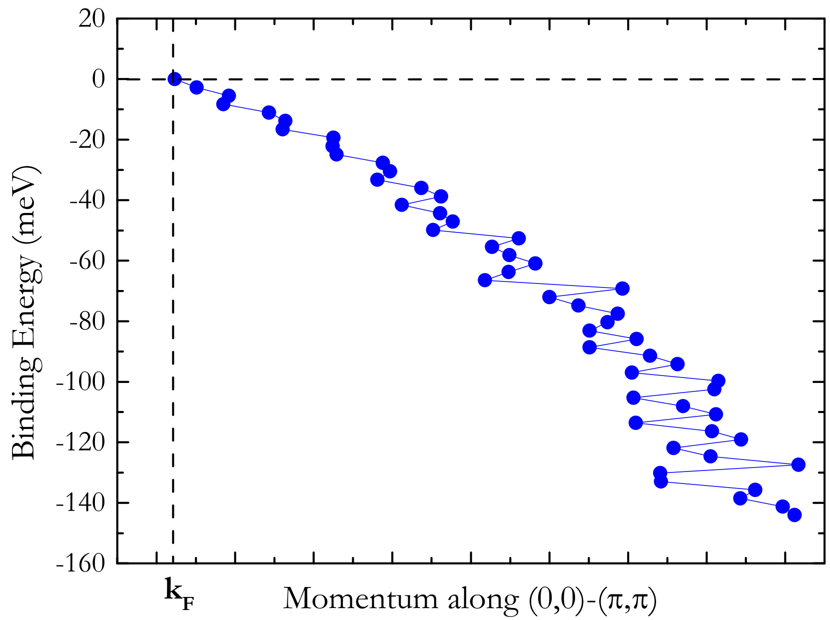
It may be an exciton-like mode

Is 2H-$TaSe_2$ similar to the HTSC?

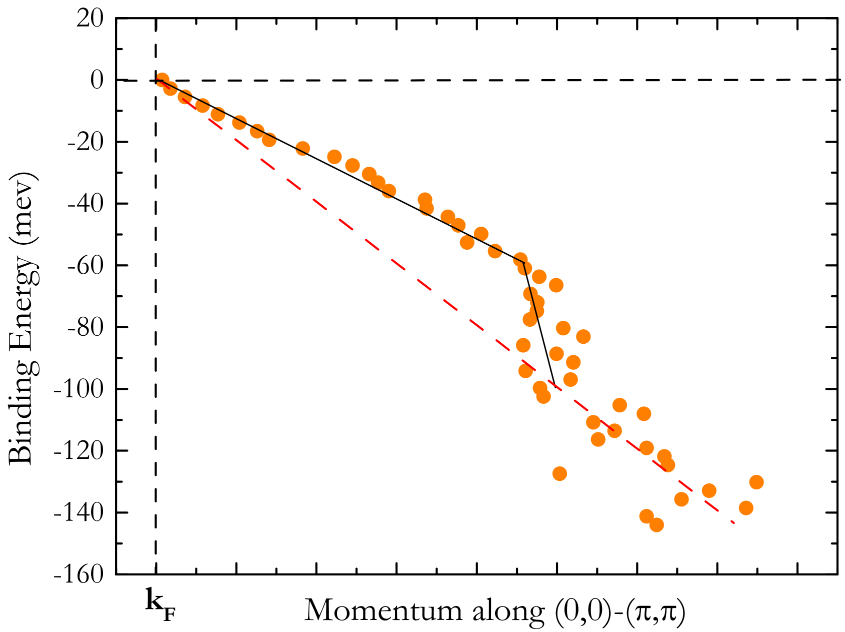
/of course not, however…/

Dispersions relations in underdoped (T_C=80 K) $Bi_2Sr_2CaCu_2O_8$
along (0,0) to (π,π) /gap node/

A. Normal State, T=120 K

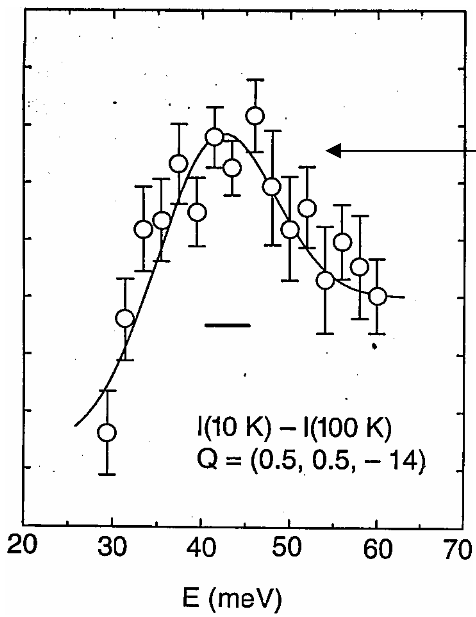


B. Superconducting state, T=45 K



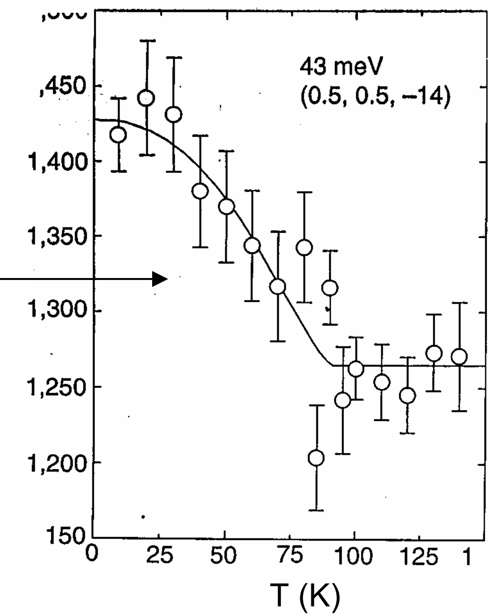
Neutron scattering from Magnetic excitations in $Bi_2Sr_2CaCu_2O_8$

/H.F. Fomg et al., Nature 398, 588 (1999)/



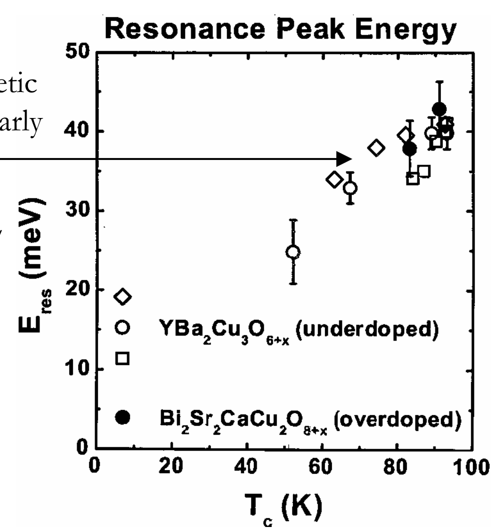
Energy profile

Temperature dependence



Energy of the magnetic excitation scales linearly with Tc

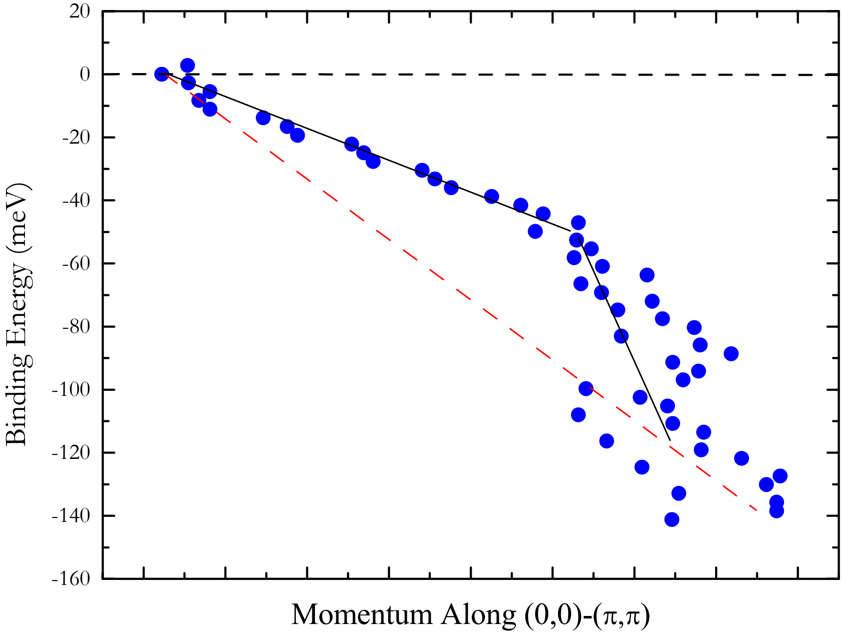
/H. He et al., cond-mat/0002013/



What will we see in ARPES?

Preliminary results on underdoped (Tc=69 K) and overdoped (Tc=51 K) $Bi_2Sr_2CaCu_2O_8$ samples

A. Superconducting State, T_c=69 K



B. Superconducting state, T_c~51 K